LA-UR-12-26380

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Title: Corrosion and Stress Corrosion Cracking: Recommendations for Mitigation and Advanced Detection

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Intended for: Report

Corrosion and Stress Corrosion Cracking: Recommendations for Mitigation and Advanced Detection

Fuel Cycle Research & Development

Prepared for
U.S. Department of Energy
Used Fuel Disposition

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10 August 2012

FCRD-XXXX-2012-XXXXXX

SUMMARY

Corrosion of stainless steel used fuel canisters in dry storage could, under certain conditions, lead to breaches of the canisters, possibly resulting in releases to the environment and introduction of moisture and oxygen into the canister, enabling possible cladding failures. With the possibility that dry cask storage may be extended to several hundred years before used fuel is disposed of in a geologic repository or reprocessed, the need to look at corrosion of the canisters containing the used fuel becomes more important. This increased importance is due to (a) corrosion mechanisms which are time dependent, with corrosion progression (not necessarily the rate) increasing as a function of time, (b) some corrosion mechanisms such as stress corrosion cracking may exhibit an incubation period, and (c) there will simply be more canisters subjected to corrosion increasing the likelihood of observing outliers.

With the possibility that during long term storage corrosion could lead to dry cask canister breach analysis is warranted to determine which corrosion mechanisms are possible and what their likelihood and consequences are, and what possible mitigation and early detection methods are available and warrant further study.

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STORAGE AND TRANSPORTATION EARLY ANALYSIS OPPORTUNITIES—STRESS CORROSION MODELING, MITIGATION, AND DETECTION TECHNIQUES

1. INTRODUCTION

Corrosion of stainless steel used fuel canisters in dry storage could, under certain conditions, lead to breaches of the canisters, possibly resulting in releases to the environment and introduction of moisture and oxygen into the canister, enabling possible cladding failures. With the possibility that dry cask storage may be extended to several hundred years before used fuel is disposed of in a geologic repository or reprocessed, the need to look at corrosion of the canisters containing the used fuel becomes more important. This increased importance is due to (a) corrosion mechanisms which are time dependent, with corrosion progression (not necessarily the rate) increasing as a function of time, (b) some corrosion mechanisms such as stress corrosion cracking may exhibit an incubation period, and (c) there will simply be more canisters subjected to corrosion increasing the likelihood of observing outliers.

With the possibility that during long term storage corrosion could lead to dry cask canister breach analysis is warranted to determine which corrosion mechanisms are possible and what their likelihood and consequences are, and what possible mitigation and early detection methods are available and warrant further study.

Used fuel dry cask storage consists of a canister containing the used fuel and a concrete cask to protect the canister and reduce worker dose. The canister is fabricated of stainless steel with longitudinal welds along the side to form the cylinder and circumferential welds to attach the bottom and perhaps to extend the height of the cylinder. The top is either welded or sealed with a gasket arrangement. Upon loading the canister with used fuel, the canister is closed, dried, evacuated and backfilled with helium. The canister then acts as the primary containment for the used fuel and provides protection for the cladding to prevent cladding degradation. The canister is then placed in a concrete cask (either vertical or horizontal) that acts to protect the canister from physical damage and to reduce worker dose rates.

While stainless steel is fairly corrosion resistant, under certain conditions, it can corrode at a relatively high rate, particularly in weld heat affected zones. Since the concrete cask is not sealed, and in fact designed to promote airflow for heat removal, water and moisture can contact the canister. Furthermore, in coastal regions, there is a possibility of deposition of salt aerosols. These salt deposits can absorb moisture and result in liquid brine in a process known as deliquescence. These deliquescent brines can have high ionic strength and be particularly corrosive.

This is not to say that corrosion will happen to the extent to result in breaches to the used fuel canisters, but it is a possibility. Without sufficient environmental definition (temperature, liquid quantity and composition) corrosion environments can be postulated that would result in canister breach. Therefore, it is of utmost importance that the possible corrosion environments be defined. These environments must be bounding, without being overly so. If the bounding environments are too aggressive, no amount of corrosion modeling/testing will provide sufficient information to demonstrate the viability of long-term interim storage.

2. CORROSION MECHANISMS

Corrosion mechanisms consist of (a) dry oxidation, (b) general corrosion, (c) microbially influenced corrosion, (d) sensitization (knife line attack) and stress corrosion cracking, (e) localized corrosion, and (f) pitting. It is commonly understood that all of these mechanisms need the presence of liquid, either bulk liquid or liquid due to deliquescence.

2.1 Dry Oxidation

Dry oxidation occurs at a relative humidity less than a certain threshold, above which general corrosion occurs, thereby forming an adherent protective oxide film of uniform thickness. It is this protective oxide film, primarily Cr_2O_3 that results in the low corrosion rates of stainless steels. Dry oxidation rates are quite low particularly for temperatures of interest for dry cask storage [1]. No analysis is recommended for dry oxidation.

2.2 General Corrosion (passive corrosion)

General corrosion (or passive corrosion) is the uniform thinning of a metal when the metal oxide passive film is stable. When the passive film is stable, corrosion occurs slowly [2,3]. This thinning results from the formation and slow dissolution of the protective passive film on the metal surface. General corrosion can occur under immersed conditions or when a liquid film exists on the surface. As long as the passive film remains stable, the general corrosion rates will be slow and if it were to crack or spall off it would heal in a short time span[4,5].When aggressive environments break down the passive film, localized corrosion may occur.

General corrosion rates are typically dependent upon (a) temperature, (b) solution chemistry, and (c) time. Corrosion rates are temperature dependent because the corrosion rates are determined by the transport properties of reacting species in the passive film and the rate of activation-controlled ion transfer at the film-solution interface. This temperature dependence typically follows an Arrhenius relationship [6]. This relationship is typically unique for a given chemistry. The solution chemistry general corrosion rate dependence is due to the fact that as the passive film becomes less stable, reacting species can transport through the film more readily [4]. While corrosion scientists can estimate which environments will be more aggressive (e.g. high chloride, low pH, low NO3-) modeling these rates is problematic without appropriate experimental data. As general corrosion progresses the rate tends to decrease. This is due to the increased passive film thickness that results in longer transport times for the reacting species [4].

<u>Recommended analysis</u>: Considering the relatively slow general corrosion rates of stainless steels, it is recommended that a review of corrosion literature be performed to arrive at a reasonable bounding corrosion rate for temperatures relevant to interim storage. These rates will inform the program whether an extensive general corrosion program is necessary or whether it can be shown that even using bounding solutions that general corrosion rates will be slow enough that general corrosion can be discounted as a concern for interim storage for time frames of several hundred years.

2.3 Stress Corrosion Cracking (SCC)

Stress corrosion cracking is the process by which cracks initiate in a material under tensile stress in the presence of a corrosive environment [7]. These cracks can grow fairly readily and could result in breaches of the canisters in interim storage. The stress corrosion process is influenced by environmental conditions, including temperature, oxygen availability, ionic concentrations, and stress profiles[7-9]. The cracks can initiate on existing flaws, or on apparently smooth surfaces (often referred to as incipient cracks) [10]. In the absence of cyclic stresses (as would be found in interim storage) it is generally observed that initiation of stress corrosion cracking does not occur on a smooth surface if the surface stress is below a threshold stress [11] and will arrest if a sufficient stress profile is not maintained [12]. Stress corrosion cracking has been modeled using the slip dissolution and film rupture model that relates the crack growth rate to the periodic rupture of the metal oxide film, dissolution, and repassivation process at the crack tip.

<u>Recommended Analysis</u>: It is recommended that an analysis be performed on relevant environments to determine the crack growth rate for a given environment and the residual stress profile. This information could be used to determine that stress corrosion cracking is not a reasonable mechanism to breach the canister during interim storage and/or what the timeframe is for breach from the onset of initiation. Since

the time for onset is not known, crack growth rates could inform the minimum frequency with which canisters should be inspected for cracks. Alternatively, this information could also inform methods for possible stress corrosion cracking mitigation.

2.4 Localized Corrosion

Localized corrosion is a phenomenon in which corrosion progresses at discrete sites or in a non-uniform manner [2]. At least upon initiation, the propagation rate of localized corrosion is faster than that of general corrosion. This increased corrosion rate is caused by the passive film being compromised allowing for the aggressive environment to contact the bare metal surface. Localized corrosion can happen either under a crevice (crevice corrosion) or in the form of pitting corrosion. Pitting corrosion requires harsher conditions to initiate than for crevice corrosion initiation [13].

Models for localized corrosion consist of an initiation and propagation components. Localized corrosion initiation requires that the open-circuit corrosion potential (E_{corr}) exceed or equal a critical potential ($E_{critical}$). $E_{critical}$ is defined as the potential below which a passive film will be stable on a metal surface (i.e., the passive film will reform spontaneously if damaged) and above which localized corrosion is possible. Both E_{corr} and $E_{critical}$ are highly dependent upon solution chemistry (e.g. pH, $Cl^–$, $NO_3^–$) and temperature [2]. There is little controversy over the initiation models for localized corrosion. However, there is some debate about the best way to experimentally determine $E_{critical}$. For crevice corrosion, the form of the crevice matters. It has been shown that crevices with Teflon tape are more aggressive than metal/metal crevices.

The propagation component of localized corrosion is subject to debate. Some corrosion scientists argue that the rate decreases with time (sometimes referred to as stifling) [14], while others argue that as the corrosion propagates it can make the crevice more severe and that the rate stays at least constant.

<u>Recommendations</u>: 1. It is recommended that a bounding but reasonable bulk solution chemistry be determined. With this information, for interim storage relevant environments, it should be possible to show that these bulk corrosion solutions are not aggressive enough to facilitate localized corrosion (except possibly at welds, see section on Sensitization). This approach would in effect argue that localized corrosion cannot initiate for relevant environments. 2. It is recommended that bounding but reasonable deliquescent brines and brine volumes be determined. It is possible that these brines could be aggressive enough to initiate localized corrosion. If that is the case then experiments and modeling will need to determine what happens as the corrosion progresses. This would require data to support stifling arguments and or demonstrate that the brine solution is consumed during corrosion, in effect stopping the corrosion behavior.

2.5 Microbially Influenced Corrosion (MIC)

Microbially Influenced Corrosion is the contribution to the corrosion of a metal or alloy due to the presence or activity, or both, of microorganisms [2]. MIC most often occurs due to the increase in anodic or cathodic reactions due to the direct impact of microorganisms on the alloy or by indirect chemical effects on the surrounding solution. Microorganisms can affect the corrosion behavior of an alloy either by acting directly on the metal or through their metabolic products. For example, some aerobic bacteria may produce sulfuric acid by oxidizing reduced forms of sulfur and thereby increase the corrosivity of the environment [15,16]. Most microbes can thrive only when the relative humidity is above 90%, although some can be active at a relative humidity as low as 75% [17].

<u>Recommendations</u>: The environment within the cask systems should be analyzed to determine if microbial activity could be supported. Until it is shown that the environment within a cask could support microbial activity, no corrosion work or analysis is necessary

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2.6 Sensitization (Knife Line Attack)

Sensitization, also known as knife line attack, is a form of intergranular corrosion of an alloy, such as stabilized stainless steel, along a line adjoining a weld that resulted in heating the material into the sensitization range (~700°C) [18]. Although sensitization might be considered a manifestation of SCC, it has aspects which merit additional discussion. In the sensitization range, carbon that was bonded with stabilizers (e.g. titanium, niobium) interacts with chromium forming chromium carbides at the grain boundaries. This interaction acts to lower the effective chromium content in the protective oxide, making the material more susceptible to corrosion. The zones can also act as local galvanic couples, causing localized galvanic corrosion. Sensitization can be overcome by proper heat treatment, which depending upon the alloy requires heating the entire part to ~1000°C followed by rapid quenching [19]. This approach of course would not be possible for canister closure welds. It was, however, demonstrated on a full size waste package prototype for the Yucca Mountain Project.

Recommended Analysis: A review of the fabrication methods of dry storage canisters should be performed to determine if the materials may be subjected to sensitization and if the welding process could result in sensitization. If found to be the case, possible mitigation methods (e.g. different materials, heat treatment, spray coatings) should be analyzed.

3. MITIGATION METHODS

Depending upon the corrosion environment and the form of corrosion several possible mitigation methods are available.

3.1 Lower Storage Temperatures

By lowering the environmental temperature all forms of corrosion would become less significant. However, this approach would result in significantly higher storage costs as fewer fuel rods could be stored in a given canister. This method is not recommended for further analysis.

3.2 Canister Cleaning

If salt build up leading to possible deliquescent brines, or microbial activity was determined to be of concern, canisters could be periodically cleaned to reduce the quantity of available salts and remove microbe masses. This method would result in operational issues, increase operating costs and worker doses. This method is not recommended for further analysis at this time.

3.3 Spray Coatings

It is possible to spray coat welds and other areas particularly susceptible to corrosion with a metallic coating. These coatings could take the form of a sacrificial coating such as aluminum that would be consumed prior to the canister material. This would be analogous to zinc sacrificial anodes used on ships and bridges. Alternatively, the coating could be corrosion resistant to protect the welds from corrosive environments. This would in effect move the problem to the edge of the spray coating, but if the base metal was sufficiently corrosion resistant then this would mitigate weld sensitization and other concerns such as stress corrosion cracking at welds. The coatings would only have to be effective while the canister was relatively hot as corrosion behaviors drop as the temperature drops. It is recommended that spray coatings receive a detailed analysis to determine the benefits and feasibility of applying them to new and/or existing canisters.

3.4 Burnishing

Since stress corrosion cracking can only occur in the presence of tensile stresses, such as residual stresses in a weld, it is possible to prevent SCC by inducing compressive stresses. If a compressive stress of several mm is induced, then no SCC can occur until corrosion has removed the compressive layer.

Burnishing can be accomplished either through low-plasticity burnishing or laser peening. Low plasticity burnishing consists of rolling a large tungsten carbide ball across the surface with considerable downward force. Several mm of compressive stress can be induced in this fashion. Laser peening can also induce several mm of compressive stress[7]. Peening is accomplished by repeatedly pulsing a laser on the surface, usually with a damper such as water to cause the force to react inward. This technique is used to induce compressive stresses on turbine blades to extend fatigue limited lifetimes and is approved by the Federal Aviation Administration. It is recommended an analysis be performed to consider the benefits and feasibility of applying burnishing to new and/or existing canisters.

4. DETECTION METHODS

Ease of inspection with the possibility of early detection of corrosion has the potential advantage of reducing the burden to demonstrate that corrosion will not occur for hundreds of years on used fuel canisters. Perhaps if it can be demonstrated that the most aggressive corrosion mechanism will not affect canisters for say 100 years, then every 50 years or so an inspection could be performed. Unfortunately, current detection methods—particularly for cracks as in SCC—are limited in their ability to be performed in situ while a canister is within a cask. However, there is mounting evidence that non-linear acoustic techniques could be introduced into a cask through ventilation holes, for instance, and inspect welds or other areas of concern with considerable sensitivity and high signal to noise ratios, especially for detecting cracks. The technique has already demonstrated the ability to see fatigue cracks that are not observable with other techniques and this sensitivity should be applicable to SCC cracks as well. An overview of all detection methods—with an emphasis on SCC—is instructive and helps place these nonlinear methods in context. A description of these nonlinear methods then follows. Based upon research to date, it is recommended that further analysis of these nonlinear methods be performed.

4.1 Detection Challenges

There are two main ways stress corrosion cracking takes place, across grains (transgranular or TGSCC) and along grain boundaries (intergranular or IGSCC). Interestingly, intergranular SCC (IGSCC) was not a design concern for early boiling water reactors.[20] However, the mechanism of IGSCC has now been evaluated in detail and various measures for preventing and mitigating IGSCC have been established.[21] Both forms of SCC have a brittle-like appearance and brittle behavior; however, metal with severe SCC can still appear bright, shiny, and perfectly “normal”, even though filled with microscopic cracks. Moreover, many materials affected by SCC do not usually display abnormal mechanical properties (e.g., yield strength and tensile strength) making ultrasonic detection challenging.[22] Thus, it is not uncommon to miss SCC in inspections and it often goes undetected until failure.

SCC is usually divided into initiation and propagation phases. In reactors (especially on the primary water side of Pressurized Water Reactors (PWR) for example) the initial phase of SCC in certain alloys may only take a few years to develop to a point where rapid crack propagation and failure occurs. Even during the propagation phase, stress corrosion cracks grow with little to no macroscopic plastic deformation until failure [23] making SCC difficult to detect in both phases. However, in more benign environments—like what might be found in nuclear storage scenarios—SCC may often take several decades to initiate before the rapid propagation phase (and potential failure) occurs.

To avoid potential SCC issues, stabilized stainless steels are often used. Alloys that are most susceptible to IGSCC are ones that have been thermally sensitized or cold worked. (See Knife Line Attack discussion above.) Thermal sensitization can be mostly counteracted by using stabilized stainless steels; stainless steel castings and welds are generally thought not to be susceptible to IGSCC.[24] Indeed, SFST-ISG-5 (NRC 1998b) states that the “NRC staff has found that casks closed entirely by welding do not require monitoring."

4.2 Additional Concerns

One particular form of intergranular SCC may prove to be of concern to very long term storage scenarios. For areas or welds that experience large neutron fluence (flux integrated over time), even stainless steels have been shown to become susceptible to SCC. This form of SCC is irradiation-assisted SCC or IASCC. Stainless 304, 316L, 316CW and 347 have been studied and all are susceptible to IASCC.[25,26] Generally IASCC is highly dependent on neutron flux-exposure time and there is often a threshold associated with the material. Generally, these threshold values are extremely high. Sweden designates any stainless exposed to more than 10^{20} n/cm^2 as "high risk", requiring regular and frequent inspections.[22] It is generally believed that fluence levels like that Swedish "high risk" limit will never be attained in any storage scenario. Yet, there are very few studies of IASCC at extremely long times and at lower neutron flux levels. So, while it is probably a good assumption to claim that IASCC may not be a concern for storage, to rule it out for long-term storage without further investigation may not be prudent.

It has also been long thought that aqueous environments are essential for SCC. Yet, there have been recent papers published which suggest that a liquid/aqueous environment is not absolutely essential: "… the aqueous environment appears to accelerate initiation and propagation but is not a necessary condition according to this research result" [27]. Furthermore, in coastal regions, there is a possibility of deposition of salt aerosols. These salt deposits can absorb moisture and result in a liquid brine in a process known as deliquescence. These deliquescent brines can have high ionic strength and be particularly corrosive [28,29]. There was also a study reported where a series of parts were exposed to such a salt aerosol or "fog" (Southwest Research Institute [30]) and all the stainless parts under test showed evidence of SCC. Dry storage casks may simply not stay entirely "dry" for very long extended periods.

4.3 Current SCC Detection Techniques in Industry

We begin by simply listing all the techniques that have been used for SCC detection, both historically and those currently in use, as well as those methods in the R&D stages. Further explanation follows. Extra detail will be provided for the newer R&D methods, interesting since they are emerging technologies and potentially useful for detecting SCC in storage scenarios.

- Visual inspection,
- Dye penetrant,
- Eddy current (both conventional and pulsed),
- Infra-red (IR) Thermography coupled with
- Vibro- or sonic-IR techniques,
- Ultrasonic Testing/Imaging, phased array technology,
- Acoustic Emission monitoring, and
- Nonlinear Acoustics testing.

Visual inspection, either directly or via remote or guided cameras (e.g., borescopes) has been used extensively in all industries where SCC is a problem. However, as already noted, visual inspection alone without other techniques often misses the early onset of SCC so visual is always paired with other detection schemes where possible. Dye penetrant methods are still used for certain inspection scenarios but require direct access to the suspected SCC area and do not work well on closed cracks or cracks found below the surface. Magnetic flux leakage methods are limited to ferrous materials (e.g. pipelines) and do not work for stainless parts or welds. Eddy Current Testing (ECT) methods, both conventional and pulsed, are very effective and still widely used for a wide variety of SCC detection problems. Moreover, pulsed ECT methods have been applied when seeking SCC under insulation where "direct" contact of the Eddy current probe is impossible. In contrast to dye penetrant methods, ECT can see cracking below the surface as well. However, generally having access to the area under test is essential for effective ECT; pulsed ECT at a standoff distance can yield erroneous results. Current state-of-the-art detection and promising new detection techniques are highlighted next.

4.3.1 Acoustic Techniques, Phased Arrays

In spite of the cost effectiveness and simplicity of the methods above, ultrasonic methods using phased arrays for flaw detection have become state-of-the-art. The OlympusNDT OmniScan portable system coupled with a number of different phased arrays have been used to locate and image individual cracks as well as colonies of SCC.[31] NDT Phased arrays, like similar ultrasound arrays used for medical imaging, can scan an acoustic beam and paint an image of an area/sector and offer the possibility of using longitudinal and transverse waves as well, giving different images of the area. Phased array systems are, in fact, used at Los Alamos for a number of different weld inspection problems and for general flaw detection as well. A good example of imaging SCC with an Olympus phased array is shown in a screen shot of Fig. 1 taken from the paper cited above [31]. It is worth noting that the sizes of the cracks seen in this example are on the order of millimeters. As with many of the techniques already mentioned, use of a phased array requires direct access to the test area, albeit with some flexibility in positioning that is gained by the ability to steer the beam of the phased array. There is also a notable example of imaging SCC in a weld in that paper as well.

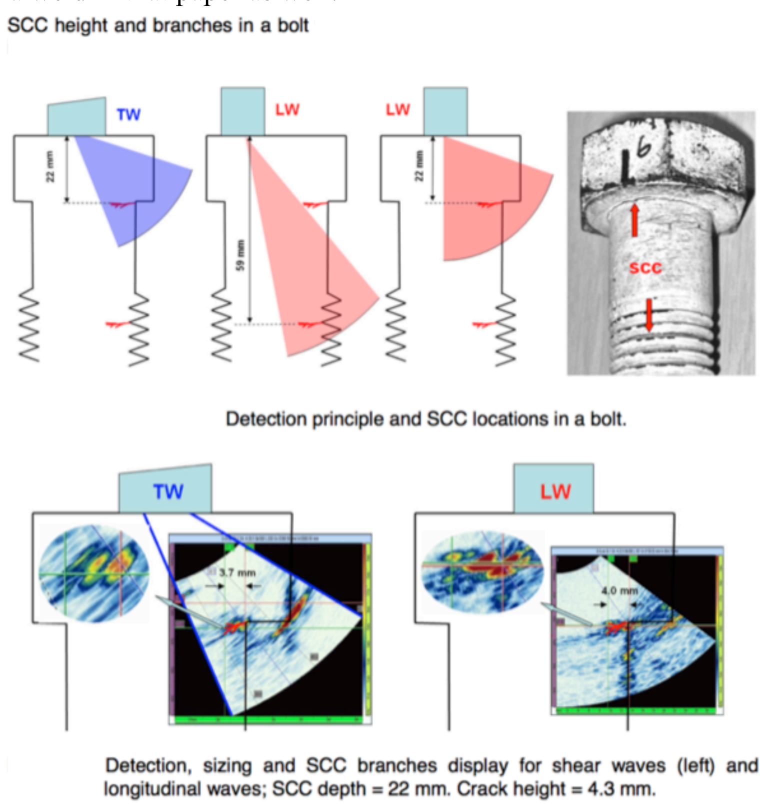
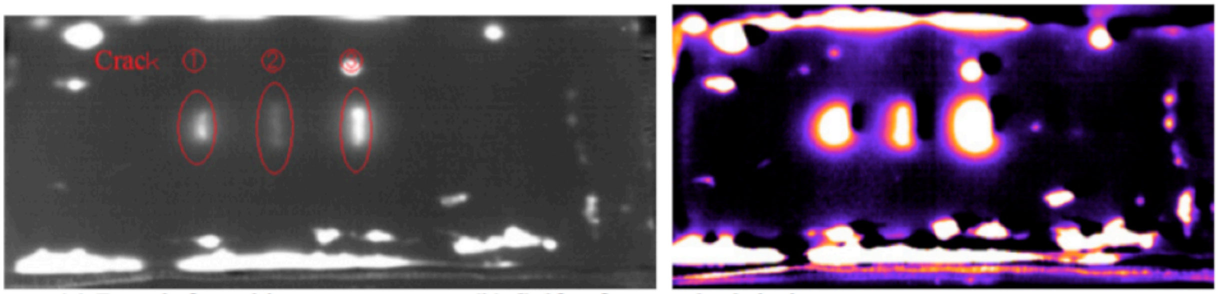


Figure 1. Demonstration showing the use of a phased array and resulting images produced from SCC seen on a common bolt.

4.3.2 Acoustic Techniques, Vibro-Infrared Imaging

Another “acoustic” technique often used to detect SCC, one that is moving from the R&D stage to development and prototyping stages, is vibro-infrared or sonic-infrared imaging.[32] The idea is to excite the area of interest with acoustic vibration and then image the area with a sensitive IR camera. Cracks will warm due to the acoustic excitation and light up the cracks on the image. A sample with 3 stress corrosion cracks was excited acoustically and the IR camera captured the resulting image shown in Fig. 2 below — taken from Fig. 4 of the paper cited above [32]. While numerous scholarly papers have been presented and published on the topic and technique, sonic-IR techniques are by no means mainstream at this time.



(a) Raw infrared image (b) Self-reference lock-in image

Figure 2. Images of crack tips taken with a sonic-IR imaging camera. Above from Ref. [14]

4.3.3 Nonlinear Acoustic Techniques

Nonlinear acoustic (NA) NDT techniques have been used in a number of applications, but unlike Sonic-IR, NA techniques have begun to move out of the R&D stage into general use and acceptance. Generally NA techniques look either for harmonic distortion of an excitation frequency or for the amount of mixing products (i.e., sum and difference frequencies) of a low frequency vibration + a small higher frequency probe wave. These two waves will interact nonlinearly in a crack or a colony of cracks like those found in SCC areas. The latter mixing is analogous to intermodulation distortion in electronic circuits. Together these techniques have been called NEWS, Nonlinear Elastic Wave Spectroscopy.[33]

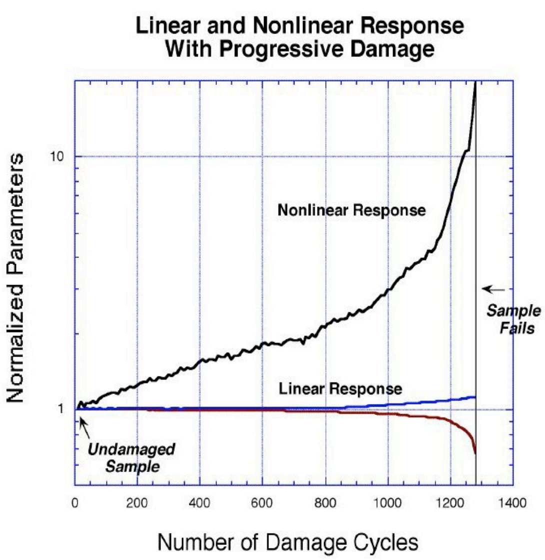


Figure 3. Comparison between progressive damage detection with linear and nonlinear methods.

NEWS techniques advance beyond standard acoustic/ultrasonic techniques to enhance the ability to detect, locate and image mechanical damage (e.g., cracks, delaminations, disbonds) at the earliest stages of development and provide quantitative metrics for monitoring continued damage accumulation. Figure 3 illustrates the extreme sensitivity of nonlinear techniques over current acoustic/ultrasonic methods for detecting and monitoring progressive damage. While this data was taken from testing performed on a plastic beam, similar results are available for a vast number of other materials including concrete, sintered metals, high temperature super-alloys, ceramics, glasses, rocks and human bone to name a few, demonstrating the breadth of application of NEWS methods.

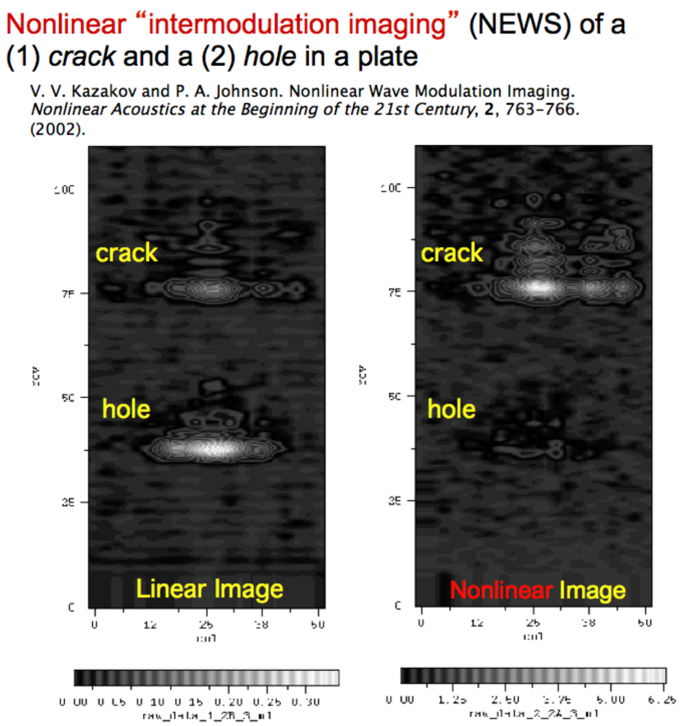


Figure 4. Imaging of a crack and a hole with linear and nonlinear imaging techniques. Note how much better the crack is illuminated in the image with a nonlinear technique.

Perhaps one of the first notable results hinting at the value of NEWS techniques was a report of an experiment showing how much better a crack was imaged with nonlinear vs linear ultrasound techniques.[34] A figure from that paper is shown below. The nonlinear image was taken by plotting the energy in the sidebands (the mixing products) around a small probe frequency wave while the plate was vibrated at a lower frequency in a manner similar to the Sonic-IR experiment discussed above. The hole in the nonlinear image (right hand side) is completely invisible and together the two images form a complementary picture of the two flaws in the plate.

Another example of NEWS imaging is shown below.[35] A poorly made diffusion bond between two disks was imaged with standard linear ultrasonic techniques and with a nonlinear technique. Again, the nonlinear technique showed areas where cracks were prevalent, places that the standard acoustic technique failed to pick up anything significant. We mention this particular example because the vibration was applied locally and remotely to each scan point using *acoustic time reversal*. Time reversal techniques could theoretically be used to propagate sound via a ventilation port access to almost any desired point (e.g. the weld). This last point will be revisited in the Discussion section. TREND here stands for Time Reversal Nonlinear Elastic Diagnostic. The image was essentially created with a nonlinear NEWS technique together with time reversal to accurately place the excitation/vibration point of the scan.

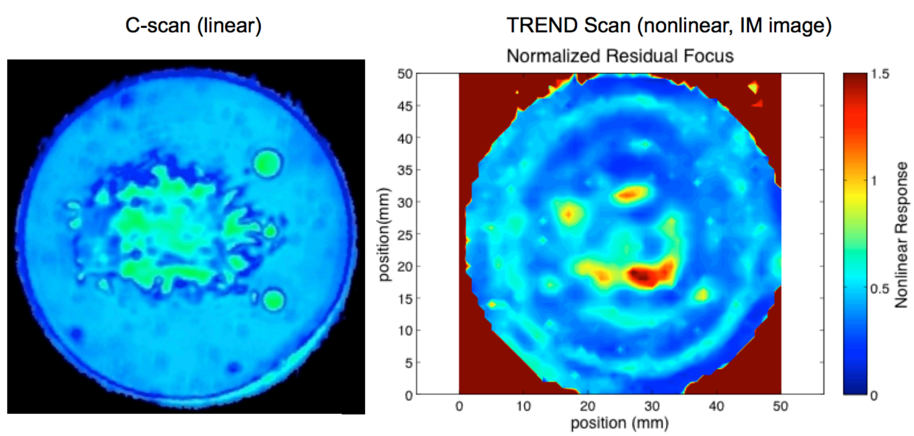


Figure 5. Comparison of linear and nonlinear images of a poor diffusion bond. The linear image is only sensitive to voids, the nonlinear image detects cracks and delaminations.

In a detection experiment more closely resembling what might be encountered when searching for SCC, these same nonlinear acoustic techniques were used to detect the amount of fatigue cracking in bent stainless steel tubes.[36] The cracking was *not* visible and too small to see using an optical microscope yet the nonlinear technique (mixing of two frequencies) was able to distinguish between several unknown and control samples, all filled with various densities of tiny cracks. As an aside, nonlinear techniques like these are quite sensitive indicators for crack detection. A nonlinear resonance measurement on a superalloy (Haynes 230) [37] detected cracks and disbonds around the carbides on the order of 10 microns.

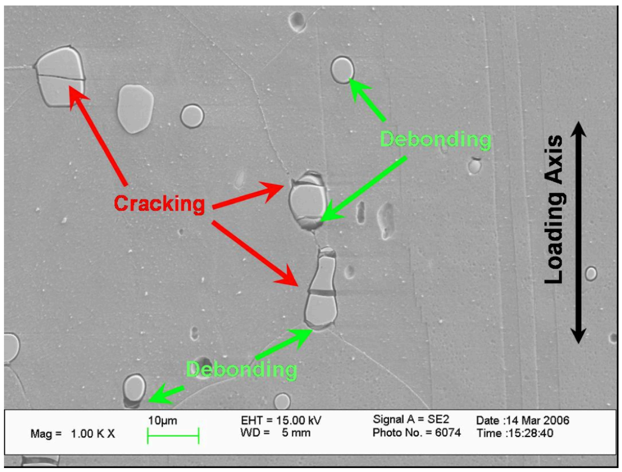


Figure 6. Micro-cracking detected around carbide inclusions in aircraft superalloy Haynes 230.

Finally, very recent nonlinear acoustics research [38,39] in Japan has been reported, motivated by detecting SCC for Nuclear Power Plants using nonlinearly generated subharmonics produced by the "clapping" of the crack. The unique signature of open and closed crack tips can give a very accurate indication of depth and size of the cracks below the surface. Ultimately the research is meant to give an accurate assessment of the amount and density of SCC and whether the crack tips are open or closed.

Finally, we highlight nonlinear acoustic techniques that have been used to track degradation of concrete itself, which has potential for monitoring the outer cask for storage. In a study of the effectiveness of "bunker busters", Nonlinear techniques were used to evaluate the integrity of a concrete bunker at increasing distance from ground zero, comparing the results to standard linear acoustic methods. The results from this study clearly illustrate the increased sensitivity of these nonlinear methods to the integrity of the concrete after the explosion, and the ability to provide a quantitative measurement for the integrity of the concrete. Other concrete tests have been performed, including the "healing" of concrete through chemical reactions with CO_2, for both carbon sequestration and marine environments.

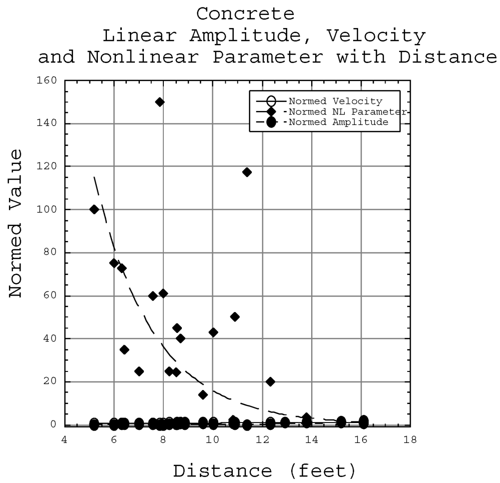


Figure 7. Nonlinearity (top curve) is a far more sensitive indicator of damage than linear methods

4.4 Discussion of SCC Detection Techniques

Almost all of the SCC detection techniques described above require direct access to the area of suspected SCC to be effective. Phased arrays can detect cracks at a standoff distance but generally the source array needs to be positioned reasonably close to the area being examined and the array needs to in direct contact with the area as well. Both sonic-IR and the Nonlinear Acoustic techniques can be made at standoff distances by applying the crack-exciting vibration with a non-contact source. The IR camera or a laser vibrometer is then used to record the image or resulting nonlinear surface vibrations at reasonable standoff distances where necessary.

Time reversal techniques could possibly be used to further advantage in either IR themography or nonlinear acoustic methods. It may prove possible to excite areas inside the cask with transducers placed via ventilation ports. If a remote camera or borescope can gain access to areas of interest, then a laser vibrometer probe accessing the same entry/exit point might open up other possibilities for detecting SCC in addition to visible inspection. Time reversal techniques were used to produce the image of the imperfect diffusion bonded disks show earlier; the sound was actually focused in the area between the disk “sandwich”. Time reversal techniques have also been used to focus sound at a point deep within a structure, to places inaccessible by any other means. [40] The figure shows how sound can be focused at a spot in a scan area between two large blocks; the clear glass block was chosen to demonstrate how well the time reversal process actually works. Amplitude on the scan area is shown in 3D and colored according to signal strength, red indicating high amplitude. So, even if direct physical access to the interior of a storage cask is difficult, time reversal offers ways probe areas otherwise difficult to access.

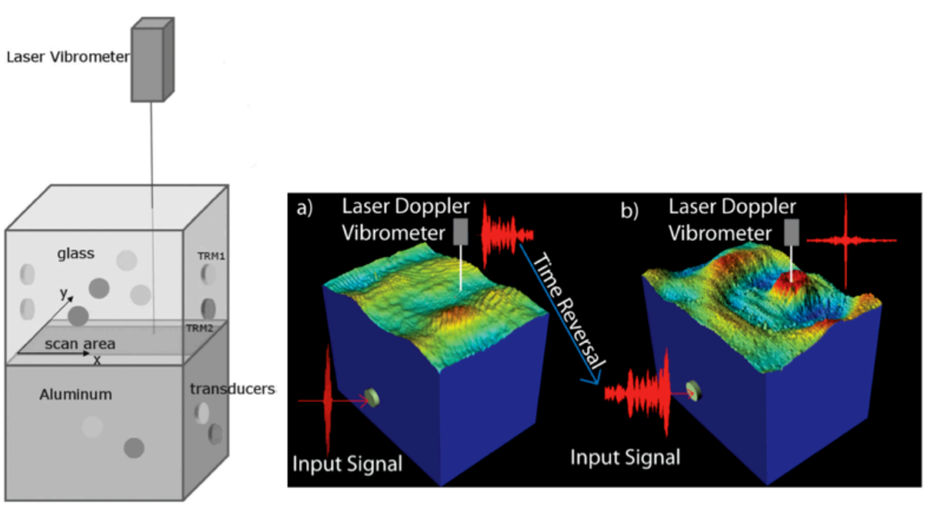


Figure 8. Using time reversal techniques to focus sound at any point and making possible the measurement of the nonlinear response at *just* that point.

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